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MINIREVIEW

Impact of a possible future global hydrogen economy on Arctic stratospheric ozone loss

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The potential role of molecular hydrogen (H_2) as a future alternative energy carrier has generated widespread interest. The possible amount of additional hydrogen emission into the atmosphere in a hydrogen-based economy depends on future hydrogen production and leakage rates throughout the complete process chain. However, the expected emissions are highly uncertain. Based on the current literature an upper limit is estimated. Additional hydrogen emissions yield enhanced water vapor concentrations in the stratosphere which will have an impact on stratospheric temperatures and on polar ozone loss. Both stratospheric water vapor and ozone are important drivers of climate change. The potential environmental risks are described and assessed to be low compared to the environmental benefits.

1 Introduction

The motivation behind a switch to "green" hydrogen as a major element of the future energy supply is to significantly reduce the emission of greenhouse gases such as carbon dioxide (CO_2) and methane (CH_4). However, H_2 leakages caused by a possible future global hydrogen economy[1–8] could yield a significant increase in stratospheric water vapor concentrations.[9] An increase of stratospheric water vapor has, in turn, an impact on surface climate and the stratospheric ozone layer, which is of societal relevance. The possible impact of a future hydrogen economy on the stratosphere therefore needs careful consideration.

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Possible impact of a future hydrogen economy on the stratospheric water vapor budget

The primary impact of a future hydrogen economy is an enhancement of the stratospheric water vapor concentration. The current abundance of water vapor in the stratosphere is the result of a complex interplay of transport, microphysics, and chemistry. The transport of air masses from the Earth's surface and troposphere (from the surface up to ≈10 km altitude) into the stratosphere (altitudes of ≈10 km to ≈50 km) occurs according to global atmospheric transport mechanisms. The net exchange between the troposphere and the stratosphere is associated with the large-scale Brewer–Dobson circulation.[10,11] The air enters the stratosphere through the cold tropical tropopause and is "freeze dried" down to a few ppmv of water.[12] After crossing the tropical tropopause, the air ascends into the upper stratosphere and finally descends at polar latitudes within a typical transit time of 4 to 6 years from the Earth's surface to the upper stratosphere (see Fig. 1).[13] The lower stratosphere tends to be very dry whereas in the middle and upper stratosphere water vapor mixing ratios increase due to the oxidation of methane (the main *in situ* source of stratospheric H_2O).

Broader context

A hydrogen-based economy seems to be environmentally friendly because the emissions of greenhouse gases would be reduced significantly as defined in the long-term goal of the "Kyoto Protocol". Therefore molecular hydrogen (H_2) has the potential to become a substantial future energy carrier provided that hydrogen is produced from renewable energy sources. Possible atmospheric consequences, in particular for the stratosphere such as stratospheric cooling or the impact on polar ozone loss, are the subject of current discussions. In this minireview, risks for the stratosphere are assessed as most likely to be low compared to potential environmental benefits for climate change and air quality.

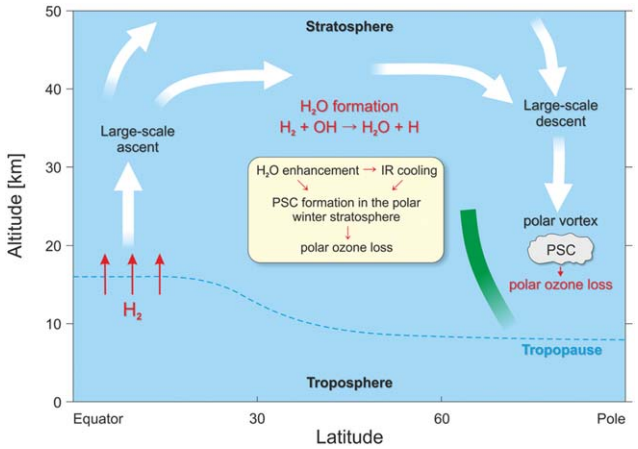

Fig. 1 Impact of H_2 emissions on the stratosphere, in particular on polar ozone loss (for details, see Section 1). The white arrows indicate the large-scale transport of air masses in the stratosphere associated with the Brewer–Dobson circulation. The edge of the polar vortex is marked in green.

Molecular hydrogen oxidation also contributes to the *in situ* production of water vapor in the stratosphere.[9] In contrast to water vapor, the cold tropical tropopause does not represent a barrier for the hydrogen and methane molecules contained in the air parcels ascending into the stratosphere. As a result, the relative importance of H_2 and CH_4 for the stratospheric water vapor budget is greatly enhanced (compared to the moist troposphere). Additional anthropogenic hydrogen release in the troposphere may therefore result in a significant increase of stratospheric water vapor.[14,15]

Consequences of enhanced stratospheric water vapor for surface climate and ozone–climate interactions

Changing the amounts of greenhouse gases in the Earth's atmosphere affects climate by altering the radiative balance between incoming solar radiation and outgoing infrared (thermal) radiation. The term 'forcing' is used to indicate that the Earth's radiative balance is pushed away from its normal state.[16] Water vapor is the most important greenhouse gas and an increase of its stratospheric concentration contributes to tropospheric warming and stratospheric cooling.[17] However, there has been a debate about the magnitude of the radiative effects.[16,18–23] There is some evidence of a sustained long-term increase in stratospheric water vapor of around 0.05 ppmv yr^{-1} from 1980 until roughly 2000. Since then the water vapor concentrations in the lower stratosphere have decreased.[16,24–26] It has been suggested that this trend has persisted for the last 40 years of the 20th century.[23] Forster and Shine[23] calculated the atmospheric temperature response to changes in stratospheric water vapor over the last 20 years of the 20th century using observed water vapor trends.[16,24–26] The associated forcing since 1960 is about 40% of the forcing due to the well-mixed greenhouse gases, or about 75% of the forcing due to CO_2 alone.[23] In a more recent study, Solomon *et al.*[27] (2010) concluded that such long-term variations in stratospheric water (time scales of decades) represent an important driver of decadal global surface climate change. Long-term variations (or trends) in stratospheric water also play an important role for ozone–climate interactions.

In addition to stratospheric cooling, enhanced water vapor has the potential to affect stratospheric ozone depletion, in particular in the polar regions.[19,28–30] Enhanced stratospheric water vapor would lead to a more frequent formation of polar stratospheric clouds (PSCs) that consist either of solid or liquid particles.[31–33] PSCs are responsible for chlorine-induced polar ozone loss *via* heterogeneous reactions that yield the production of activated chlorine from the reservoir substances.[32] Consequently, enhanced PSC formation results in more efficient polar ozone loss. Therefore enhanced water vapor would have the potential to delay the recovery of the stratospheric ozone layer expected due to the phase-out of ozone-depleting substances regulated under the "Montreal Protocol" and its subsequent Amendments and Adjustments.[34]

A prerequisite for the formation of the Antarctic ozone hole in spring is a cold polar vortex that isolates polar from mid-latitude air masses in the wintertime stratosphere. The Arctic polar vortex is less stable, warmer, smaller, and more variable than the Antarctic vortex yielding, in general, less and more variable ozone depletion in Arctic spring. The reason for this difference is a stronger planetary wave activity in the northern hemisphere caused by the different distribution of land masses in the Antarctic and Arctic.[11]

In the polar Arctic vortex, temperatures are near the threshold for chlorine activation in contrast to the Antarctic where, in general, the temperatures are much lower (by about 10 K).[34] Therefore, the period of PSC occurrence is much shorter in the Arctic (up to $\approx$3 months) than in the Antarctic (5 to 6 months) and no PSC formation occurs in Arctic warm winters.[34,35] Temperatures during Arctic winters depend critically on the dynamic conditions of the specific winter[36] and severe halogen-driven ozone loss occurs only for sufficiently cold winters. Despite uncertainties in the future development of temperatures and dynamic conditions of the polar Arctic vortex,[35,37] increases in stratospheric water vapor will allow activation in the Arctic vortex to occur when it would otherwise not.[28] Therefore enhanced stratospheric water vapor is a crucial factor for Arctic ozone loss and will be discussed in detail within this paper.

In the following, the impact of a future hydrogen economy on the stratosphere is assessed based on the current literature. First, in Section 2, possible additional H_2 surface emission rates caused by a potential future hydrogen economy will be discussed to obtain a reliable estimation of the increase in the tropospheric H_2 concentration. In Section 3, the impact of such an increase on stratospheric water vapor and related ozone–climate interactions will then be discussed.

2 Impact of enhanced H_2 emissions on tropospheric hydrogen budget

2.1 Tropospheric hydrogen budget

To assess the impact of a future hydrogen economy on the stratosphere, in particular on water vapor and ozone, which are both relevant for surface climate, the impact of hydrogen on the tropospheric H_2 budget must be examined.

Currently, molecular hydrogen has an average mixing ratio of about 530 ppbv in the troposphere.[38] The major sources of H_2 are fossil fuel use, biomass burning (including biofuel), nitrogen fixation, photochemical production from CH_4, and other volatile organic compounds. The reaction of H_2 with OH and soil uptake are the major sinks of H_2 in the troposphere. The strength of each term is given in the literature[38–44] and compiled in Ehhalt and Rohrer[44] (2009). The current production (76 Tg yr^{-1}) and loss rates (79 Tg yr^{-1}) yield a tropospheric H_2 lifetime of 2 years.[44,45] Today about 50% of the total tropospheric H_2 production can be considered anthropogenic.[44] The average vertical distribution of H_2 in the lower and middle stratosphere proves to be relatively uniform as in the troposphere.[44]

Due to the long lifetime of H_2 in the troposphere, the local emissions of H_2 spread out in the troposphere. Therefore, additional hydrogen emissions yield an increase of the mean tropospheric H_2 mixing ratio determining the entry of H_2 into the stratosphere through the tropical tropopause. Because molecular hydrogen is oxidized to form water vapor in the stratosphere,[9] any additional hydrogen affects stratospheric temperatures, the development of stratospheric ozone, and the climate.

2.2 Possible future hydrogen production and emission scenarios

Hydrogen is an energy carrier which can be produced by a variety of methods (*e.g.* electrolysis, coal and biomass gasification, or steam reforming) using renewable, fossil, or nuclear power.[46] The amount of additional future hydrogen emissions into the atmosphere has to be estimated on the basis of the expected future production and leakage rates throughout the whole process chain. Therefore the process steps of production, conditioning, transportation, storage, and consumption of hydrogen have to be considered carefully by applying a so-called life cycle assessment.[5,8]

However, the production and utilization rates of hydrogen in a future global energy system are difficult to forecast today. Both parameters will strongly depend on future political decisions and on technological developments in the end-user area (*e.g.* highly efficient, reliable, and cost-effective fuels cells for automotive and stationary applications). Today, the magnitude of emissions due to hydrogen leakages can only be estimated since real measurements for different hydrogen technologies are only partly available in the literature. In addition, the development of hydrogen-related technologies is still ongoing, and this will help to reduce hydrogen losses even further.

One of the first studies to discuss a possible impact of a future hydrogen economy on the atmosphere was published by Tromp *et al.*[3] (2003). They assume a scenario according to which all current technologies based on oil or gasoline combustion are replaced by hydrogen fuel cells and imply an overall hydrogen leakage rate of 20%. These assumptions would lead to anthropogenic H_2 emissions that raise the concentration of H_2 at the Earth's surface to 2.3 ppmv (about four times the current global annual mean of 0.53 ppmv). This study has been assessed to be unrealistic due to the high leakage rate assumed.[1,2,4,6,47]

In contrast, Warwick *et al.*[4] (2004) used only a 5% leakage rate, but the same assumption about the production rates. A scenario proposed by Schultz *et al.*[2] (2003) assumes that only 50% of current fossil-fuel-based technologies will be replaced by hydrogen and estimate a leakage rate between 3% and 10%. The latter scenario is regarded as more realistic compared to the previous scenarios and indicates that H_2 at the Earth's surface is raised to 1.1 ppmv.[2,6]

To estimate a reliable magnitude of future hydrogen production in a hydrogen-based economy, the future economic and population growth will have to be taken into account. A scenario considering the future hydrogen production until 2100 "IIASA-SRES B1-H2" performed by the International Institute for Applied Systems Analysis (IIASA) is based on the "IIASA-SRES B1" simulation published by the Intergovernmental Panel on Climate Change (IPCC).[48] Baretto *et al.*[49] (2003) complemented the "IIASA-SRES B1" scenario by a potential future hydrogen economy. In this scenario, a maximum production of hydrogen (2750 Tg yr^{-1}) will be reached around the year 2080. Afterwards, hydrogen production will decrease, due to a regressive world population growth and the presence of a more efficient technology for energy conversion.

The potential role of hydrogen in energy systems with and without climate policy was analyzed by van Ruijven *et al.*[50] (2007) with a long-term energy model based on the "IIASA-SRES B2" scenario.[51] Based on current technology, they conclude that hydrogen is too expensive to become a major end-use carrier. However, it is expected that its cost will decrease as technologies significantly improve over time. They found that assumptions made by Baretto *et al.*[49] (2003) are much more optimistic on the future role of hydrogen in the global energy system in comparison to their own sensitivity studies.

The scenario "World Energy Technology Outlook (WETO-H2)"[52] performed by the European Commission describes the development of the global energy supply system until 2050 assuming a rapid implementation of a future hydrogen economy. By mid-century (2050), hydrogen production will reach 11% (349 Tg yr^{-1}) of the current global fossil fuel consumption.

A recently published paper by Bond *et al.*[8] (2011) upscaled projections for global H_2 production and consumption for direct energy and industrial end-uses until 2100 based on 2008 estimations from Suresh *et al.*[53] (2010). They estimate a production of 225 Tg yr^{-1} for 2050 and a maximum production of 1692 Tg yr^{-1} for 2100.

As has been emphasized, there are many uncertainties in the hydrogen production expected in the future. Therefore, it is only possible to estimate a reliable range of magnitude for the possible future hydrogen production taking into account socioeconomic and ecological aspects. Comparing all these production scenarios, the "IIASA-SRES B1-H2" (year 2080) scenario is clearly an upper limit.

Future possible hydrogen emissions depend on both future production scenarios and expected leakage rates. Losses can occur along the complete process chain during production, distribution, storage, and use of hydrogen. Zittel and Altmann[54] (1996) report that losses of gaseous H_2 are significantly less than 1%, while those of liquid H_2 depend on the method of handling and are in the order of 1–10%. Further studies[55,56] estimate hydrogen leakage rates throughout the complete process chain. Findings by Colella *et al.*[55] (2005) show that a gaseous-hydrogen-based economy can be designed to be low in hydrogen leakage (<3%), while in a liquid-hydrogen-based economy an unintentional loss of hydrogen may occur to the atmosphere. Van

Ruijven *et al.*[56] (2011) distinguish between chains of high and low leakages on the basis of the leakage rates at the individual points of the process chain. Chains of high leakage rates contain more liquid hydrogen, involve more trucks, and more transfusion between transport modes. Van Ruijven *et al.*[56] (2011) estimate a minimum and maximum chain emission between 0.3% and 10% depending on system configuration. The lower end of this range would in fact be equal to current emissions from fossil fuel combustion. Similar to expected future hydrogen production, only an extreme upper limit can be estimated for future expected leakage rates.

Only hydrogen emissions are relevant for studying the impact of a hydrogen-based economy on the atmosphere. The emissions are calculated from the global production and the individual leakage rates. Because of the uncertainties discussed above, in most studies the impact of a future hydrogen-based economy on the atmosphere is analyzed using scenarios reflecting low, intermediate, and/or very high (worst-case) hydrogen emission rates.[2,3,6,56,57]

Fig. 2 shows the range of projected hydrogen production rates with the "IIASA-SRES B1-H2" scenario as a clear upper limit. Hydrogen leakage rates in the range between an optimistic and a pessimistic leakage rate of 1% and 10%, respectively, are highlighted in gray. The second ordinate shows the corresponding estimated tropospheric hydrogen increase. The assumptions made in previous studies[2–4] to assess the impact of future increasing hydrogen emissions on the atmosphere are also indicated. Based on the discussion above, previous studies can be assessed as shown in Table 1.

3 Possible future impact of enhanced H_2 emissions on the stratosphere

The impact on the stratosphere of enhanced stratospheric water vapor caused by increasing H_2 emissions has only been studied by a limited number of studies using different types of atmospheric models and techniques at various levels of complexity. In the following, the details of theses studies will be discussed.

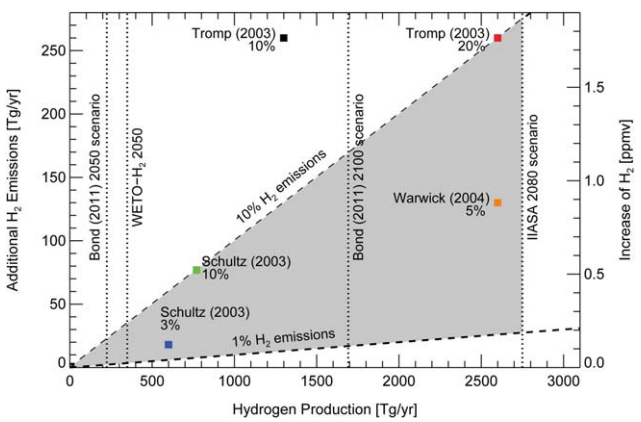

Fig. 2 Projected hydrogen production and emissions. The "IIASA-SRES B1-H2" scenario (for details, see Section 2) is a clear upper limit for future predicted hydrogen production. Additional hydrogen emissions assuming a leakage rate in the range between an optimistic (1%) and a pessimistic (10%) scenario are highlighted in gray. Assumptions made in previous studies[2–4] are marked in color (adapted from Feck[58] (2009)).

Table 1 Various scenarios for a potential future global hydrogen economy result in different tropospheric mixing ratios of molecular hydrogen and different assessments (adapted from Feck[58] (2009))

Case	Reference	H_2/ppmv	Category
Ref	Current	0.56	Reference stratosphere
S1	Schultz *et al.*[2]	0.65	Very low H_2 emissions
S2	Schultz *et al.*[2]	1.10	Low H_2 emissions
S3	Warwick *et al.*[4]	1.40	Intermediate H_2 emissions
S4	Tromp *et al.*[3]	2.30	Very high H_2 emissions

3.1 Impact on stratospheric water vapor

To infer future H_2O mixing ratios and H_2O enhancements caused by potential future H_2 emissions in the stratosphere and feedback on climate, the transport mechanisms of H_2O from the troposphere into the stratosphere have to be simulated reliably. Unfortunately, current chemistry climate models (CCMs) are limited in their representations of key processes that control the distribution and variability of water vapor within the stratosphere and do not consistently reproduce the recently observed changes in stratospheric water vapor.[27,59–61] For example, most models have a poor representation of the seasonal cycle in tropical tropopause temperatures, which control global stratospheric water vapor abundances.[60]

To avoid this problem, we refer to an alternative method used by Feck *et al.*[6] (2008) including observations to calculate the distribution of stratospheric water vapor caused by enhanced hydrogen emissions. The reference of stratospheric water vapor is based on a climatology inferred from satellite measurements by the HALOE instrument onboard the UARS satellite.[62] To calculate the additional amount of water vapor in the stratosphere caused by enhanced hydrogen emissions, the assumption is used that air masses with equal CH_4 mixing ratios encounter a comparable state of hydrogen oxidation regardless of whether they were transported poleward by the global-scale mean circulation or whether they stayed in the tropics.[6] In contrast, an increase of stratospheric water vapor caused by other sources, such as enhanced transport from the troposphere (*e.g.* due to changes in the tropical tropopause temperature) or volcanoes, results in a very different vertical profile of H_2O in the stratosphere. Based on box model studies for single ascending air parcels performed by Feck *et al.*[6] (2008), the oxidation of CH_4 and H_2 can be calculated for each methane level in the stratosphere[6,57] to infer the additional amount of H_2O (ΔH_2O) in the stratosphere from increased H_2 emissions caused by a possible future hydrogen economy.[2–4]

Using this approach, the 3-dim spatial distribution of stratospheric water vapor influenced by enhanced hydrogen emissions can be calculated from recent climatological stratospheric CH_4 and H_2O distributions for different H_2 emission scenarios S1–S4 (*cf.* Table 1). Fig. 3a shows climatological stratospheric water vapor for late Arctic winter conditions (in general, the period with the strongest Arctic ozone loss) inferred from satellite measurements by the HALOE instrument onboard the UARS satellite.[62] The additional amount of stratospheric water vapor (ΔH_2O) for scenario S2 and S4 is shown in Fig. 3b and Fig. 3c, respectively, based on the same methane and water vapor climatology inferred from HALOE measurements.[62] Fig. 3b and

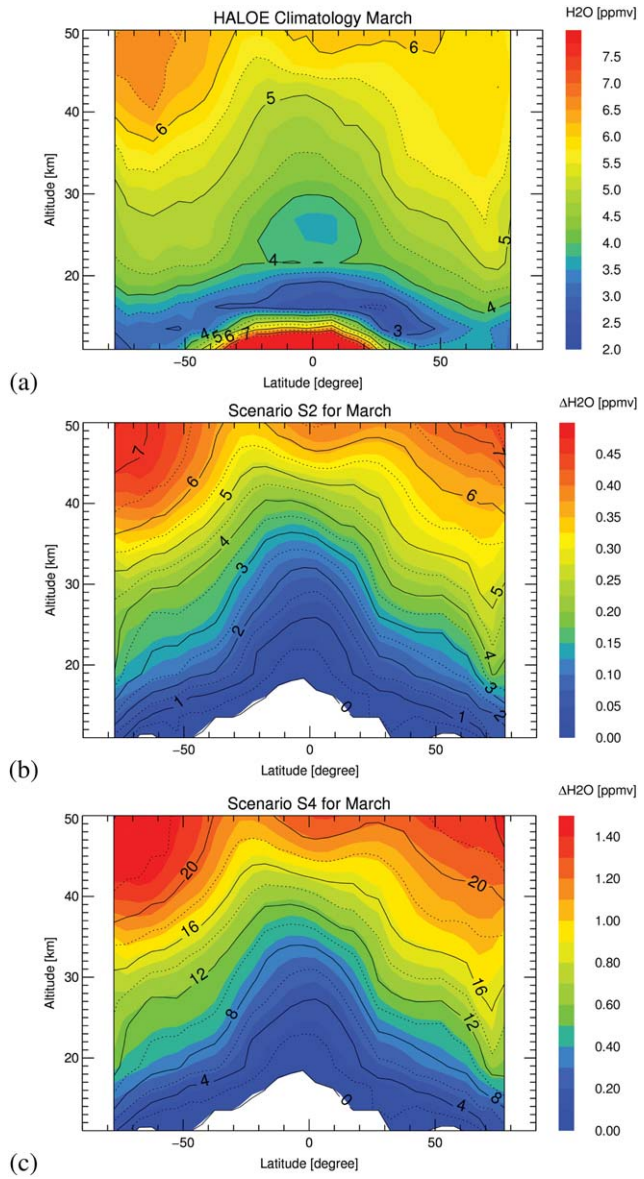

Fig. 3 (a) Stratospheric H_2O climatology derived from HALOE measurements for the month of March. (b) Calculated enhancements in stratospheric water vapor (ΔH_2O) assuming scenario S2 (for details, see text). The black lines indicate the percentage change. (c) The same as (b), but for scenario S4.

3c show that the strongest impact of potential H_2 emissions on the stratospheric water vapor distribution will be at high latitudes. The H_2O enhancements in the high-latitude lower stratosphere for scenario S2 (see Fig. 3b) are relatively low (up to $\approx$0.2 ppmv) compared to decadal changes of stratospheric water vapor in the mid-latitude lower stratosphere ($\approx$1 ppmv) observed in the last twenty years of the 20th century.[16,24–26] Scenario S4 yields up to $\approx$0.5 ppmv additional water vapor mixing ratios in the high-latitude lower stratosphere, but is regarded as unrealistic (see Section 2). Because enhanced stratospheric water vapor caused by a possible future hydrogen economy has a systematic impact on the stratosphere which is superimposed by the long-term variability of the stratosphere, there is a need for further analysis.

3.2 Impact on ozone–climate interactions

In general, increases in stratospheric water vapor act to cool the stratosphere due to greater thermal emissions. However, the upper troposphere warms from absorption of the increased downwelling thermal radiation.[17] The observed water vapor increase of around 1 ppmv in the lower stratosphere during the last twenty years of the 20th century[16,24–26] contributes significantly to the observed cooling of the lower stratosphere.[18–23] In simulations, Forster and Shine[23] (2002) calculated a cooling of about 0.8 K in the tropical lower stratosphere and up to 1.4 K at high latitudes for a 1980 background stratosphere for a homogeneous stratospheric water vapor increase of about 1 ppmv in the lower stratosphere.[16,24–26] However, it was shown that a computed stratospheric temperature change caused by increasing stratospheric water vapor is strongly dependent on the temporal and spatial variation of stratospheric water vapor changes.[22,63,64] A more realistic change in the stratospheric water vapor profiles yields a weaker stratospheric cooling than for a constant profile.[22,63,64] In particular, Myhre *et al.*[64] (2007) calculated a stratospheric cooling of about 0.0 K to 0.2 K in the lower polar stratosphere ($\approx$15–25 km) caused by stratospheric water vapor increase due to CH_4 oxidation based on observational data for the time period from 1979 until 2000. The amount and the spatial distribution of stratospheric water increase calculated by Myhre *et al.*[64] (2007) are comparable to the water vapor increase inferred for a potential future hydrogen economy (see Fig. 3b, scenario S2).

The major impact of stratospheric water vapor distribution on future H_2 emissions is expected in the polar regions as shown in Fig. 3 (*cf.* Section 3.1). Feck[58] (2009) calculated the expected decrease in stratospheric temperatures for the Arctic polar vortex assuming an increase of stratospheric water vapor according to scenarios S2 and S4 using the fixed dynamical heating approximation[65] implemented in the two-dimensional radiative transfer model RAD.[66] This model has been validated against results calculated by the radiative transfer model used by Forster and Shine[18,23] (1999, 2002). Based on RAD simulations, Feck[58] (2009) calculated a mean temperature change in scenarios S2 and S4 of -0.1 K and -0.2 K, respectively, for the polar vortex region ($\approx$16–26 km). These results are consistent with the range of global average temperature changes due to a hydrogen economy mentioned by Warwick *et al.*[4] (2004). In contrast, Tromp *et al.*[3] (2003) estimated a cooling of 0.5 K per 0.5 ppmv H_2O based on Forster and Shine[23] (2002) (*cf.* above). However, as discussed above, the findings by Shine *et al.*[63] (2003) and Myhre *et al.*[64] (2007) show that a constant positive offset of the stratospheric water vapor mixing ratios as used in Forster and Shine[23] (2002) yields, in general, a stronger cooling than a variable change of stratospheric H_2O. Therefore, the stratospheric cooling in Tromp *et al.*[3] (2003) is overestimated.

The calculated stratospheric cooling in the polar lower stratosphere calculated by Feck[58] (2009) for scenario S2 and calculated by Myhre *et al.*[64] (2007) for CH_4 oxidation alone are based on similar values for stratospheric water vapor increase and yield a comparable stratospheric cooling. In lower latitudes, the temperature change caused by a possible future hydrogen economy is smaller than in polar regions according to the spatial distribution of the water vapor enhancements. Feck[58] (2009)

calculated a change in the mean annual stratospheric temperature of about −0.04 K and −0.1 K for scenario S2 and S4, respectively.

Thus, the stratospheric cooling caused by a potential future hydrogen economy calculated by Feck[58] (2009) is in the range of the stratospheric cooling caused by methane oxidation alone for the last twenty years of the 20th century calculated by Myhre *et al.*[64] (2007). In addition, it should be noted that the observed water vapor increase of around 1 ppmv in the lower stratosphere in the last twenty years of the 20th century is too large to be attributed to methane oxidation alone.[16,24–26] Thus, further mechanisms are proposed that can be linked to other external forcing agents or are related to changes in tropopause temperatures and circulation.[16] A stratospheric water vapor increase caused by a potential future hydrogen economy is most likely of minor importance for stratospheric cooling compared to other stratospheric water vapor sources. However, there could be feedback in the system that we currently do not understand.

An important remaining question is whether enhanced water vapor values caused by a future hydrogen economy have the potential to significantly affect halogen-driven ozone loss processes, in particular in the Arctic. Hitherto, different types of atmospheric models and techniques at various levels of complexity ranging from ozone loss proxies,[6] 2-dimensional models[3,4] or 3-dimensional chemistry transport models[56,57,67] have been used to determine the impact of enhanced stratospheric H_2O caused by an increased atmospheric H_2 burden on ozone loss processes in the Arctic. Ozone loss proxies[6] are based on an empirical relationship between the Arctic ozone column loss and the total volume of air exposed to temperatures below a critical value controlling PSC formation.[37,68,69] Atmospheric 2-dimensional models use the strong zonal symmetry component of the atmospheric circulation and calculate the distribution of chemical trace gases as a function of the two dimensions of latitude and altitude (or pressure). In addition, a longitudinal dependence is considered in 3-dimensional chemistry transport models using wind fields from meteorological analyses provided by weather service centers.

In previous studies, results range from significant impacts[3] to results concluding that an increase in molecular hydrogen emissions is unlikely to have a substantial impact on polar stratospheric ozone.[4,6,57] Because the H_2 leakage rate deduced by Tromp *et al.*[3] (2003) was assessed to be unrealistically high,[1,2,4,6,47] the impact of a potential hydrogen economy on the stratospheric ozone loss is obviously overestimated. The findings of other studies[4,6,56,57,67] vary somewhat as a result of using different models and techniques. However, all these studies suggest that a future potential hydrogen economy would have a minor impact on Arctic ozone loss.

We present some details to give an idea of the magnitude of the potential ozone loss caused by a future hydrogen economy. Typical values for the total column ozone range between 200 DU–400 DU (Dobson Unit). In cold Arctic winters, an accumulated ozone loss was found of up to approximately 100 DU at the end of the winter.[70] The chemical ozone destruction over the Arctic in early 2011 was, for the first time in the observational record, comparable to that in the Antarctic ozone hole with a maximum accumulated ozone loss of up to approximately 130 DU.[71] In a study recently published by Vogel *et al.*[57] (2011) using the 3-dimensional chemistry transport model CLaMS,[72–74] an additional accumulated ozone loss for the very cold Arctic winter 2004/2005 (the coldest Arctic winter on record since 1960[75]) was calculated. Maximum values of 2.5 DU additional ozone loss were found using a realistic scenario for hydrogen emissions (S2) and current chlorine levels. This will become even smaller as chlorine levels are expected to decline in the next few decades. This value of 2.5 DU is equivalent to 4% of the total simulated column ozone loss of 65 DU over the Arctic winter 2004/2005. It should be noted that the cooling of the stratosphere caused by the stratospheric water vapor enhancements was also considered within this study.[57] Increasing H_2O concentrations according to scenario S4 ("worst case scenario") have the potential to affect polar ozone loss processes during the cold Arctic winter 2004/2005 up to 6.8 DU ($\approx$11%) of additional accumulated Arctic ozone loss.[57] For both scenarios S2 and S4, the calculated ozone loss is much smaller than the existing variability of ozone loss observed in cold Arctic winters.[70]

It should be noted that simulations focusing on the stratospheric impact of additional hydrogen economy differ from each other not only in the model or method used, but also in the assumptions related to chemical or physical processes. Tromp *et al.*[3] (2003) consider only the change in stratospheric water vapor without considering the impact of reduced fossil fuel emissions such as carbon monoxide, methane, or nitrogen oxide on the stratosphere. The impact of reduced fossil fuel emissions is also not investigated in studies performed by Feck *et al.*[6] (2008) and Vogel *et al.*[57] (2011). This is the case because the focus of these studies is the impact of a possible future hydrogen economy on Arctic polar ozone loss, where changing water vapor concentrations and temperatures are the critical parameters for halogen-driven ozone loss. The reduction of atmospheric pollutants is additionally implemented in studies performed by Warwick *et al.*[4] (2004), Jacobson *et al.*[76] (2005), and van Ruijven *et al.*[56] (2011). Other differences are the consideration of aerosol particle change, radiative forcing, and the time periods of the simulations. Therefore the results of the studies differ depending on processes that are considered in model simulations. However, in summary, it can be concluded that the studies discussed above using different models, methods, and assumptions show that overall the risks for polar stratospheric ozone are most likely small in contrast to the early studies performed by Tromp *et al.*[3] (2003).

4 Discussion and conclusions

Previous studies[2,4,56,67,76] show that positive impacts for climate, air quality and health caused by the reduction of emissions from fossil fuels such as greenhouse gases (aim of the "Kyoto Protocol") and many pollutants such as nitrogen oxides or sulfur dioxide would occur using hydrogen as a major element of the future energy supply. The expected benefit depends critically on the method used for production and storage of hydrogen, in particular, renewable energy sources such as wind-powered electrolysis should benefit the climate most.[5,7,56,67,76]

However, enhanced emissions of molecular hydrogen caused by a potential future hydrogen economy would yield increasing water vapor concentrations in the stratosphere, in particular at high latitudes. Future increasing stratospheric water vapor is an

important driver of climate change. The expected stratospheric water vapor increase caused by a possible future hydrogen economy is a systematic change of stratospheric water vapor due to additional emissions of hydrogen. This increase and its impact on both stratospheric cooling and stratospheric polar ozone loss is estimated to be small compared to the variability of stratospheric water vapor values. According to our current knowledge, the potential risks for the stratosphere, in particular for polar regions, are most likely negligible using reliable estimates of future hydrogen emissions and considering the expected decreasing stratospheric chlorine loading in the next few decades as a result of the "Montreal Protocol". Provided that hydrogen is produced from renewable energy sources, the environmental benefits and the minor risks for the stratosphere reinforce the conclusion that hydrogen as an energy carrier is a reasonable alternative to fossil fuels.

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